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Incipient plasticity in metallic thin films

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The authors have compared the incipient plastic behaviors of Al and Al–Mg thin films during indentation under load control and displacement control. In Al–Mg, solute pinning limits the ability of dislocations to propagate into the crystal and thus substantially affects the appearance of plastic instabilities as compared to pure Al. Displacement control allows for a more sensitive detection of such instabilities, as it does not require collective dislocation motion to the extent required by load-controlled indentation in order to resolve a yield event. This perception is supported by *in situ* transmission electron microscopy observations. © *2007 American Institute of Physics*.
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The onset of macroscopic plastic deformation during indentation is thought to correspond to the first deviation from elastic response in the load versus displacement curve. For load-controlled indentation of crystalline materials, this deviation commonly has the form of a displacement burst at constant indentation load. The elastic shear stress sustained prior to this excursion is often much higher than predicted by conventional yield criteria.[1] The physical origin of the enhanced elastic loading and the subsequent displacement burst has been the subject of extensive discussions in literature;[2] however, many researchers[3–6] agree that the onset of macroscopic plastic deformation is primarily controlled by dislocation nucleation and/or multiplication.

The initial yield behavior of metals is in some cases characterized by a series of discontinuous yield events rather than a single one.[4,7] Because of the characteristic steps that result from these yield events during load-controlled indentation, this phenomenon is commonly referred to as "staircase yielding." This process repeats until fully plastic loading is established.[5] Whereas extensive staircase yielding occurs in pure Al thin films, it was recently found that Al–Mg thin films show essentially continuous loading behavior under otherwise identical conditions.[8] In this letter, we report in detail on the influence of solute Mg on plastic instabilities in indentation of Al–Mg.

One pure Al film and one Al–Mg film with a Mg concentration of 2.6 wt % were evaporated onto Si substrates (see Ref. 9 for details). During evaporation, the substrates were kept at 300 °C to establish a grain size of the order of the film thickness, which was approximately 300 nm for both specimens.

Ex situ nanoindentation measurements were conducted both under load control and displacement control using a TriboIndenter (Hysitron, Inc., Minneapolis, MN) system equipped with a Berkovich indenter with an end radius of curvature of approximately 120 nm. Scanning probe microscopy was used to image the surface prior to each indentation to select a target location on a smooth flat area of the specimen away from the wedge. In both control modes, the indentation rate was approximately 10 nm/s.

In situ nanoindentation experiments under displacement control were performed in a transmission electron microscope (TEM) using a quantitative indentation stage, which has recently been developed.[10] The stage was equipped with a Berkovich indenter with an end radius of approximately 150 nm, as measured by direct imaging in the TEM. The *in situ* indentations were carried out on the Al and Al–Mg films at the cap of the wedge, where the surface has a lateral width of the order of 300 nm. The displacement rate during indentation was 7.5 nm/s.

Given the significant rounding of the indenter in both types of experiments, the initial loading is well described by spherical contact up to a depth of the order of 10 nm. In Tabor's approximation, the elastoplastic strain due to spherical loading is proportional to $\sqrt{\delta/R}$, where δ is the indentation depth and R the indenter radius;[11,12] the equivalent strain rate is therefore proportional to $1/\sqrt{(4\delta R)}\ d\delta/dt$. Using the above mentioned values it is easily seen that at a depth of 10 nm, the initial strain rates in both types of experiments compare reasonably well to one another, with values of 0.14 and 0.10 s^{-1} for the *ex situ* and *in situ* experiments, respectively.

The load-controlled indentation measurements show displacement bursts during loading on both the Al and the Al–Mg films, as illustrated in Fig. 1. The curvature of the loading portion prior to the first excursion is well described by elastic Hertzian contact, as indicated by the dashed curves. Since the depth over which the tip is rounded is larger than the depth over which the initial elastic behavior is expected, the expression for a spherical indenter is used.[13] The subsequent yield behavior is classified as staircase

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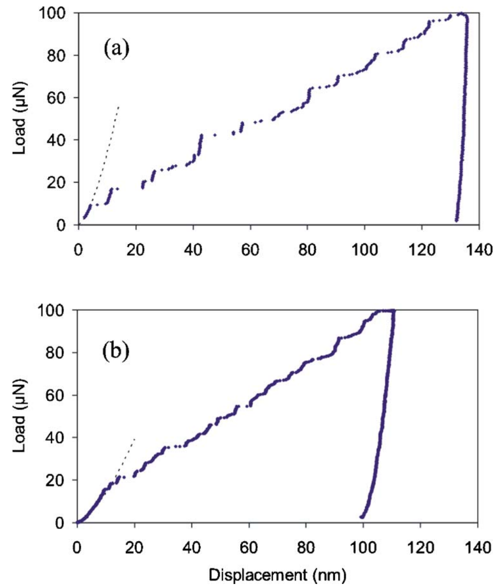

FIG. 1. *Ex situ* load-controlled indentation response of (a) pure Al and (b) Al–2.6%Mg. The dashed lines represent elastic indentation by a spherical indenter with a radius of 120 nm, with the respective elastic moduli calculated from the slope of the unloading curve.

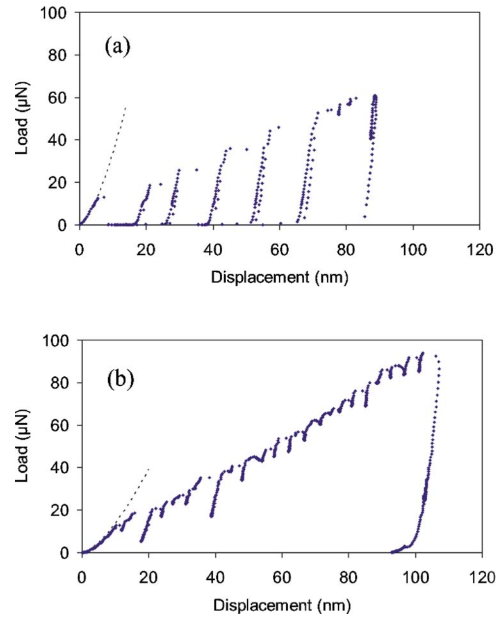

FIG. 2. *Ex situ* displacement-controlled indentation response of (a) pure Al and (b) Al–2.6%Mg.

yielding due the aforementioned dislocation-based mechanisms.[3,14]

The displacement bursts encountered in Al–Mg have a magnitude of up to 7 nm, which is substantially smaller than those observed in pure Al, being up to 15 nm in size. In earlier experiments,[8] it was observed that the attenuation of displacement bursts occurs for Mg concentrations both below and above the solubility limit in Al, from which it was inferred that the effect is due to solute Mg, which impedes the propagation of dislocation bursts through the crystal. Consequently, at a constant indentation load and for a given amount of stored elastic energy, fewer dislocations can be pushed through the solute atmosphere of Al–Mg than through a pure Al crystal, which accounts for the observed difference in size of the yield excursions. Comparison of Figs. 1(a) and 1(b) furthermore reveals that the loading portions between consecutive yield events in Al–Mg show significant plastic behavior, whereas in Al they are well described by elastic loading, which at these higher indentation depths are attested by a slope that is intermediate between spherical[13] and Berkovich[15] elastic contact. The plasticity observed in Al–Mg can be explained in terms of the solute pinning of dislocations nucleated during the preceding yield excursion.

Under displacement control, the effect of solute drag on the initial yielding behavior becomes much more evident, as illustrated in Fig. 2. The loading curves of both Al and Al–Mg show pronounced load drops, which have the same physical origin as the displacement excursions in load-controlled indentation, i.e., stress relaxation by bursts of dislocation activity. Also in this case, the loading behavior up to the first load drop closely follows the elastic response under spherical contact. However, the appearance of the load drops is very different: in pure Al, the load drops are large and mostly result in loss of contact, while in Al–Mg they are smaller and more frequent, and contact is maintained during the entire loading segment. The forward surges occurring with each load drop are a result of the finite bandwidth of the feedback system. In the case of pure Al, the observations of complete load relaxation and loss of contact are indicative of the stored elastic energy being fully released in the forward surge before the feedback system is able to reduce the load. In Al–Mg, however, solute pinning strongly reduces the dislocation velocity, which enables the feedback system to respond fast enough to maintain elastic contact. Thus, not all of the stored elastic energy is inputted back into the specimen.

The comparison between the load-controlled and displacement-controlled experiments shows that discrete yield events are far more resolvable under displacement control. This may be rationalized as follows. When the critical shear stress for a dislocation source under the indenter is reached under load control, a discernable strain burst results only if the source is able to generate many dislocations at constant load, i.e., the load-displacement curve must shift from a positive slope to an extended range of zero slope for the slope change to be readily detected. This again is possible only if the newly nucleated dislocations can freely propagate through the lattice, as in pure Al. Under displacement control, however, provided that the feedback bandwidth is sufficiently high, the system may respond to the decrease in contact stiffness when only a few dislocations are nucleated, causing a distinct shift from a positive to a steeply negative slope in the load-displacement curve. Therefore, a detectable load drop can occur without collective propagation of many dislocations and as such may easily be observed even under solute drag conditions. This result cautions against using only load-controlled indentation to determine whether yielding proceeds continuously.

The observations of incipient plasticity are illustrated in Fig. 3 by the TEM images and load-displacement data recorded during an *in situ* displacement-controlled indentation on Al–Mg (for *in situ* observations of pure Al, reference is made to Ref. 10). While the indented grain is free of dislocations at the onset of loading [Fig. 3(a)], the first dislocations are already nucleated within the first few nanometers of the indentation [Fig. 3(b)], i.e., well before the apparent initial yield point that would be inferred from the load versus

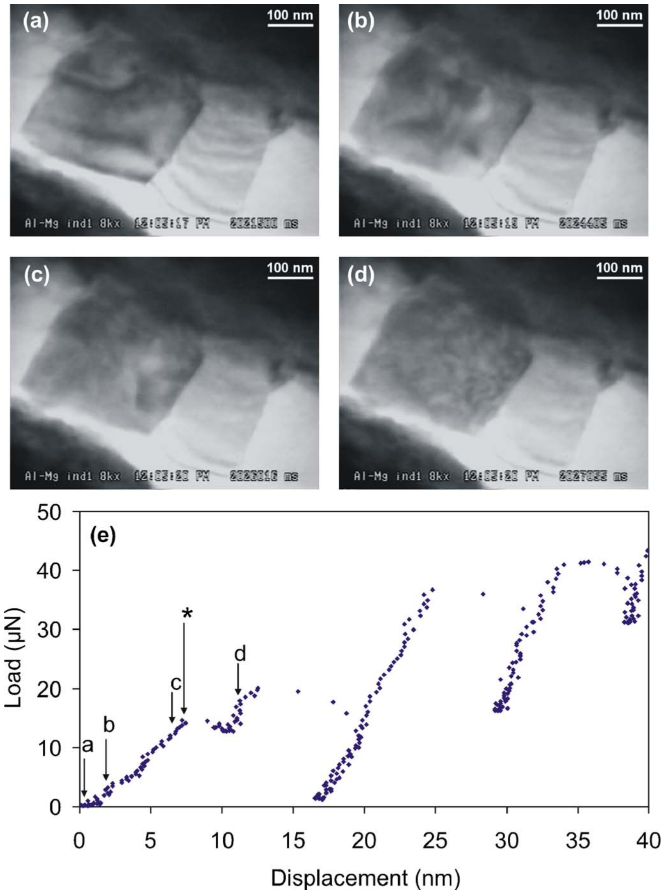

FIG. 3. TEM bright-field image sequence [(a)–(d)] from the initial loading portion (e) of an *in situ* displacement-controlled indentation on Al–2.6%Mg. The first dislocations are nucleated between (a) and (b), i.e., prior to the apparent yield point (*). Although individual dislocations cannot be easily distinguished in the video frames, their nucleation is evidenced by an abrupt change in image contrast: before nucleation, only thickness fringes can be seen, whereas more complex contrast features become visible at the instant of nucleation.

displacement data only. At the inception of the first macroscopic yield event, dislocations are present throughout the entire grain [Fig. 3(c)]. The yield event itself is associated with a rearrangement of these dislocations, which significantly changes the appearance of the dislocation structure [Fig. 3(d)]. However, the number of newly nucleated dislocations between (c) and (d) is relatively small, as also becomes clear from the limited increase in indentation depth (3 nm corresponds to approximately ten Burgers vectors). This supports our perception that only a small number of dislocations need to be nucleated in order for a yield event to be detected under displacement control, although the first dislocations nucleated between (a) and (b) do not provide an obvious signature in the load-displacement curve. In the case of *in situ* displacement-controlled indentation of pure Al,[10] the onset of dislocation nucleation/propagation coincides with a barely detectable yet unambiguous load drop that occurs well before the initial macroscopic yield event, which is further evidence of more collective dislocation motion in Al in comparison to Al–Mg.

The *in situ* observations of Al–Mg furthermore provide a self-consistency check for the dynamics of a yield event. With solute drag preventing full load relaxation, the size of a forward surge Δh is essentially determined by the dislocation velocity v and the mechanical bandwidth of the transducer f. Therefore, ignoring the drag exerted by the feedback system, the dislocation velocity may to a first approximation be estimated as $v \sim \Delta h f$, which, using $\Delta h = 7$ nm and $f = 125$ Hz yields a velocity of the order of 1 μm/s. This is of the same order as observed *in situ* for the initial dislocations in Fig. 3(b), which traversed the 300 nm film thickness in about 130 ms (four video frames at a frame rate of 30 frames/s).

It is concluded that the yield events are resolved more clearly under displacement control, particularly in the presence of solute drag. *In situ* TEM displacement show that many dislocations are nucleated prior to the initial macroscopic yield point and that the macroscopic yield event is associated with the rearrangement of these dislocations.

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